Interaction-assisted propagation of Coulomb-correlated electron-hole pairs in disordered semiconductors

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A two-band model of a disordered semiconductor is used to analyze dynamical interaction induced weakening of localization in a system that is accessible to experimental verification. The results show a dependence on the sign of the two-particle interaction and on the optical excitation energy of the Coulomb-correlated electron-hole pair.

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The problem of two interacting particles (TIP) in a random potential is an excellent paradigm for the general question of the interplay of disorder and interactions in many-body systems. First addressed in a 1990 paper by Dorokhov [1], the subject has been especially well studied since Shepelyansky's 1994 publication [2]. Considering the TIP localization length l_2, most authors [2–6] obtain an interaction-induced increase $l_2 > l_1$ over the single-particle localization length l_1 independent of the sign of the interaction, with $l_2/l_1 \sim l_1^a$ and $a = 1$ or $a = 0.65$ [6]. Here, l_1 and l_2 are measured in units of the lattice constant of a one-dimensional Anderson chain. Similar results have been obtained for TIP in a quasiperiodic chain [7]. The independence of the predicted effect on the sign of the interaction is an especially intriguing feature.

Early work approached this problem using a wide variety of theoretical techniques and focused on establishing the existence of the TIP-effect, while more recent work has dealt with quantitative details like scaling behaviour and the influence of interaction range, strength and sign [1–6,8–11].

Existing works comprise purely theoretical case studies since the model of just two particles in a single band does not correspond to any real physical situation. Obviously, experimental study is needed to promote further understanding of the TIP problem, and would put the presently rather academic discussion on a firm physical basis. Exploiting the fact that the coherent spatio-temporal dynamics of Coulomb-correlated electron-hole pairs is strongly influenced by the two-particle interaction, we show in the present paper that *the TIP localization properties should be accessible to modern ultrafast optical techniques*. The corresponding spatial and temporal scales are on the order of sub-μm and 1 ps. Our numerical studies are based on integration of the Semiconductor Bloch Equations and include no *a priori* assumptions about energy hierarchies or interaction matrix elements. The calculations were performed for a disordered 1D semiconductor quantum wire, where localization effects are most important. Despite its simplicity, this model system contains already all essential ingredients to describe the dynamics following optical excitation even in disordered systems of higher dimension [12]. The model parameters have been given values that resemble those of realistic disordered semiconductor quantum wires. Additional physical parameters (excitation energy, spectral pulse width, screening length, different masses of the two particles) allow the study of a wide variety of observable phenomena.

In our numerical calculations we investigate the spreading of an electron-hole wave packet after local excitation by an optical pulse. Here the interaction is given by the long-range Coulomb potential which, besides producing bound states (excitons) near the edges of the excitation spectrum, also correlates the electrons and holes in the pair continuum. Previous theoretical studies of the spatial-temporal dynamics of wave packets formed from excitons show that their motion is rather limited in the presence of scattering [13]. This result is recovered by our present calculations. Here we focus our interest on the dynamics of optically generated wave packets in the pair continuum. We find that the excitation conditions in the presence of particle-particle interaction influence the carrier dynamics dramatically. In addition, and in contrast to some previous claims in the literature, we find that the sign of the interaction has a pronounced effect on the spatio-temporal dynamics.

We consider a 1D array of sites i with diagonal disorder in both the valence band (vb) and the conduction band (cb). The site energies ε_{vi} and ε_{ci} corresponding to the vb and the cb, respectively, are randomly distributed over the interval $[-W/2, W/2]$ and are uncorrelated. The nearest neighbor cb levels are coupled by the tunneling term $-J^c$, the vb levels by $-J^v$. We use the Coulomb interaction in its monopole-monopole form [14] with matrix elements

$$V_{ij} = -\frac{U}{4\pi\epsilon\epsilon_0}\frac{e^2}{r_{ij}+\alpha} \tag{1}$$

which has been regularized in order to cope with the

pathological singularity in one dimension. The constant α has been chosen to be 5 times the lattice constant [15]. The total Hamiltonian is written as $\hat{H} = \hat{H}_0 + \hat{H}_I + \hat{H}_C$, where $\hat{H}_0$ describes the *vb* and *cb* band structure, $\hat{H}_I$ represents the semiclassical interaction with the electromagnetic field $E_i(t)$ in dipole approximation, and $\hat{H}_C$ defines the electron-electron interaction term [16,17].

In the following, we assume a *local* initial excitation at the central site $i = 0$, which is modeled by setting $\mu_i = \delta_{i,0}$ for the local dipole matrix element in $\hat{H}_I$. The optical polarization $p_{ij}(t)$ is obtained from the equation of motion for the polarization operator $\hat{p}_{ij} = \hat{d}_i \hat{c}_j$, which is coupled to the equation of motion of the electron and hole intraband quantities $\hat{n}^e_{ij} = \hat{c}^+_i \hat{c}_j$ and $\hat{n}^h_{ij} = \hat{d}^+_i \hat{d}_j$, respectively, where the operators $\hat{c}^+_i, \hat{c}_i$ ($\hat{d}^+_i, \hat{d}_i$) describe the electron (hole) creation and annihilation operators at site i. The equation of motion for the expectation values $p_{ij}(t)$ and $n^{e,h}_{ij}(t)$ is treated using the well-known Semiconductor Bloch Equations for $p_{ij}(t)$ and $n^{e,h}_{ij}(t)$ in the real-space representation [17]. Detailed derivations of the Semiconductor Bloch Equations are given in [14] and the textbook [18]. As we are interested in small excitation densities, we write only the equation for p_{ij} in the lowest (linear-response) order in the exciting field

$$\partial_t \, p_{ij} = -i\Big(\varepsilon^e_i - \varepsilon^h_j - V_{ij}\Big)p_{ij} + i\sum_{l=1}^{N}\big(J^e\, p_{il} + J^h\, p_{lj}\big) + i\mu_j E_j(t)\,\delta_{ij}. \tag{2}$$

Using the conservation laws $n^e_{ij} = \sum_l p_{lj}\, p^*_{li}$ and $n^h_{ij} = \sum_l p_{jl}\, p^*_{il}$ valid in this lowest order [19], we obtain the intraband quantities.

Instead of studying a rather academic localization length which describes only the asymptotic behavior of wave functions, we calculate the experimentally more relevant participation number $\Lambda(t) = (\sum_i n^2_{ii})^{-1}$. Here n_{ii} stands for either n^e_{ii} or n^h_{ii}. With the packet localized at site 0, $n_{ii} = \delta_{i0}$ and $\Lambda = 1$, while for an excitation uniformly extended over the sample of N sites, $n_{ii} = 1/N$ and $\Lambda = N$. Our calculations were performed for chains containing $N = 240$ sites. Boundary effects can easily be identified in the temporal evolution of Λ and do not play any role as long as $\Lambda < N/2$. All the data presented are free of finite-size effects.

The transform-limited optical pulse is defined by its mean energy $\hbar\omega$ and the temporal width τ of the gaussian envelope $\sim exp\{-(t/\tau)^2\}$. We define an excitation energy E_{exc} refered to the bottom of the (ordered) absorption band, i.e. $E_{exc} = \hbar\omega - E_{gap}$. All results are given for $\tau = 100$ fs, which corresponds to an energetic width (FWHM) of 22 meV.

To make contact with the previous work where two particles in a single band were placed initially at a single site, we first consider the situation of a symmetric band structure with $J^e = -J^h = 20$ meV. The absorption spectra with and without Coulomb interaction are shown in Fig. 1 for the ordered case. The peak structure near the absorption is due to the excitonic resonances. Upon changing the sign of the Coulomb interaction, the bound state resonances are shifted from the bottom to the top of the absorption spectrum.

As the dynamics of electrons and holes are the same for the assumed symmetric band structure, we restrict our discussion to the electrons. We first discuss the situation in the absence of Coulomb interaction. Fig. 2 shows the corresponding $\Lambda_e(t)$ for two different disorder parameters W after excitation by a pulse at $E_{exc} = 80$ meV. The excitation is centered in the absorption spectrum as indicated in Fig. 1. $\Lambda_e(t)$ evolves exponentially with rise time less than 1 ps. Here and below, we take the saturation value as a measure of localization. As expected, it decreases rapidly with increasing disorder. We find $\Lambda \sim W^{-1.3}$ as W is varied over the range 40 meV to 240 meV for $J = 20$ meV. A discussion of related exponents can be found in [20].

Fig. 2 contrasts the interacting and noninteracting behavior for two values of disorder and reveals three remarkable features. i) The interaction clearly leads to a reduction of the localization of the particles. We have carefully checked that the saturation value of $\Lambda_e(t)$ at long times is not due to a finite size effect; values $\leq N/2$ are fully converged with respect to the sample size. ii) While the participation number in the noninteracting situation evolves exponentially and saturates quickly ($<$ 1 ps), the interacting wave packets evolve diffusively and reach their saturation values at much longer times. iii) The sign of the Coulomb interaction ($U = \pm 1$) has virtually no influence on the propagation of the particles in this case. The same is true if we apply a very short excitation pulse which spectrally covers the whole band. The spectral position of the central pulse frequency within the band is then completely irrelevant. In this situation the excited particle pair-wave packet is initially situated exclusively at site $i = 0$. These observations are not new [2–4]. However, iii) has been questioned [6] and ii) remained unexplained.

In all cases where J^e and J^h are of comparable magnitude we find that the participation number is enhanced by the interaction. In a mean field picture, it is the temporal fluctuations of the field originating from the partner particle which destroy the coherence necessary to produce localization. This explanation in terms of a *dynamic*-correlation-induced weakening of the influence of disorder can be nicely corroborated by a number of case studies.

We note that contrary to previous statements, the independence of the sign of the Coulomb interaction is not a general feature, but is a consequence of the imposed electron-hole symmetry. In particular, displacing the central frequency of excitation pulses from the center of the absorption band, the situation changes completely.

Note that this choice of the excitation frequency corresponds to the realistic situation where electron-hole pairs are excited close to the absorption edge in semiconductors. Fig. 3 shows the participation number Λ for light electrons and heavy holes, i.e., $J^e = -2J^h = 20$ meV. The central excitation energy of the pulse is placed in the lower part of the pair continuum at $E_{exc} = 40$ meV. Results averaged over 60 realizations are shown for $W = 80$ meV and $U = 0, \pm 1$. The results are invariant under reflection of the excitation frequency through band center and simultaneous switching of the sign of the interaction. This reflects the approximate symmetry (within fluctuations in the site energy distribution) of the Hamiltonian.

It is at first sight counterintuitive that the enhancement of the participation number is larger for attractive ($U = -1$) than for repulsive ($U = +1$) interaction. This behavior can be attributed to the fact that for attractive interaction and positive masses (i.e. for excitation into the lower half of the excitation continuum) the electron-hole pair tends to stay closer together. The fluctuating field due to the accompanying particle is then more pronounced as compared to the case of repulsive interaction, where the mutually repulsive particle pair tends to be separated. Hence the *dynamic*-correlation-induced weakening of the influence of disorder is less effective for repulsive than for attractive interaction.

Completely different behavior is found for a *static* field. We consider an infinitely heavy hole, $J^h = 0$, which now produces a static field, and excitation at the (interaction-free) band center. For both attractive and repulsive interactions, the participation number is *decreased* with respect to the noninteracting case. This result is easily understood without invoking fluctuating fields since at band center electron states have maximal extent. In the presence of interaction, off-center states are admixed leading to greater confinement. The effect of the static interaction is thus opposite to that of a fluctuating field [21].

The strong retardation of the saturation in the interacting case can also be understood in our picture. Whether with or without interaction, the electron and hole wave packets spread over a range given by the single-particle levels involved in the optical transition just after the short excitation pulse. The fluctuating Coulomb field due to the partner particle then leads to an increase of the spread of the wave packets. As a consequence, the average fluctuating field acting on a given particle is reduced, which in turn tends to slow further spreading, eventually leading to the observed saturation at long times. The neglect of phonon interactions in our model is justified *a posteriori*. Fig. 2 makes it clear that the time scales between 100 fs and $\simeq 3$ ps are fully sufficient for experimental observations while near-band-edge acoustic phonon scattering occurs on longer time scales [22].

Previous work [2–4] on the TIP problem suggests a scaling of the two-particle localization length $l_2/l_1 \sim l_1^a (U/J)^2$, with $a = 1$ or $a = 0.65$ [6]. Our results for the participation number do not obey such a scaling law as far as the dependence on U is concerned. We obtain for electrons and holes, both for attractive and repulsive interaction, $\Lambda(U = \pm 1)/\Lambda(U = 0) \sim \Lambda(U = 0)^b$ with $b = 0.65 \pm 0.3$. For electrons and attractive interaction the present model predicts $\Lambda_e \sim W^{-c}$ with a larger exponent $c \simeq 2.2$ compared to $c = 1.3$ for the noninteracting case.

In conclusion, we have studied the localization of a pair of interacting particles in a situation which, in principle, is accessible to experiments. Optical excitation in the pair continuum of a disordered one-dimensional semiconductor with long-range Coulomb interaction has been considered. Starting from a tight-binding description, the temporal evolution of the participation numbers of the electron and the hole wave packets has been calculated by a direct solution of the equation of motion of the correlated material excitation within linear response with regard to the exciting laser field. The participation number increases with interaction for both attractive and repulsive interaction. We find that in general the degree of delocalization depends strongly on the sign of the interaction, in contrast to previously published predictions. The sign of the interaction becomes irrelevant (even if the masses of electrons and holes are different) only for two special situations: excitation in the center of the pair continuum, or excitation of the whole band. We have checked that this result is independent of the assumed form of the interaction and that it remains true also for the short-range interactions studied in the literature. Compared to the single-band models treated in the past, the present semiconductor model admits a richer variety of phenomena, which can be qualitatively explained within a mean-field picture. We emphasize that the enhancement of the participation number is clearly not due to a finite size effect, and that it should be experimentally observable. Ultra-short time-of-flight experiments on arrays of semiconductor quantum wires in the coherent limit using pump-probe techniques are a promising option. The enhancement should also be observable in disordered semiconductor quantum wells. In this case we expect the enhancement to be even more pronounced, since, in contrast with one-dimensional systems, only states close to the band edge are essentially affected by the disorder in two dimensions, so that the interaction will lead to coupling with rather extended states.

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FIG. 1. Absorption spectrum of the ordered chain for $U = 0, +1, -1$ and for equal electron and hole masses ($J^e = -J^h = 20$ meV). The spectrum of the optical pulse with $E_{exc} = 80$ meV and $\tau = 100$ fs is also given.

FIG. 2. Temporal evolution of the participation number Λ after excitation by a 100 fs pulse for the interacting (solid lines) and noninteracting (dotted lines) cases. The disorder parameters are $W = 20$ meV and $W = 60$ meV

FIG. 3. Temporal evolution of Λ_e (upper traces) and Λ_h (lower traces) for excitation in the lower half of the continuum and $U = 0, \pm 1$. $J^e = -2J^h = 20$ meV, $W = 80$ meV.

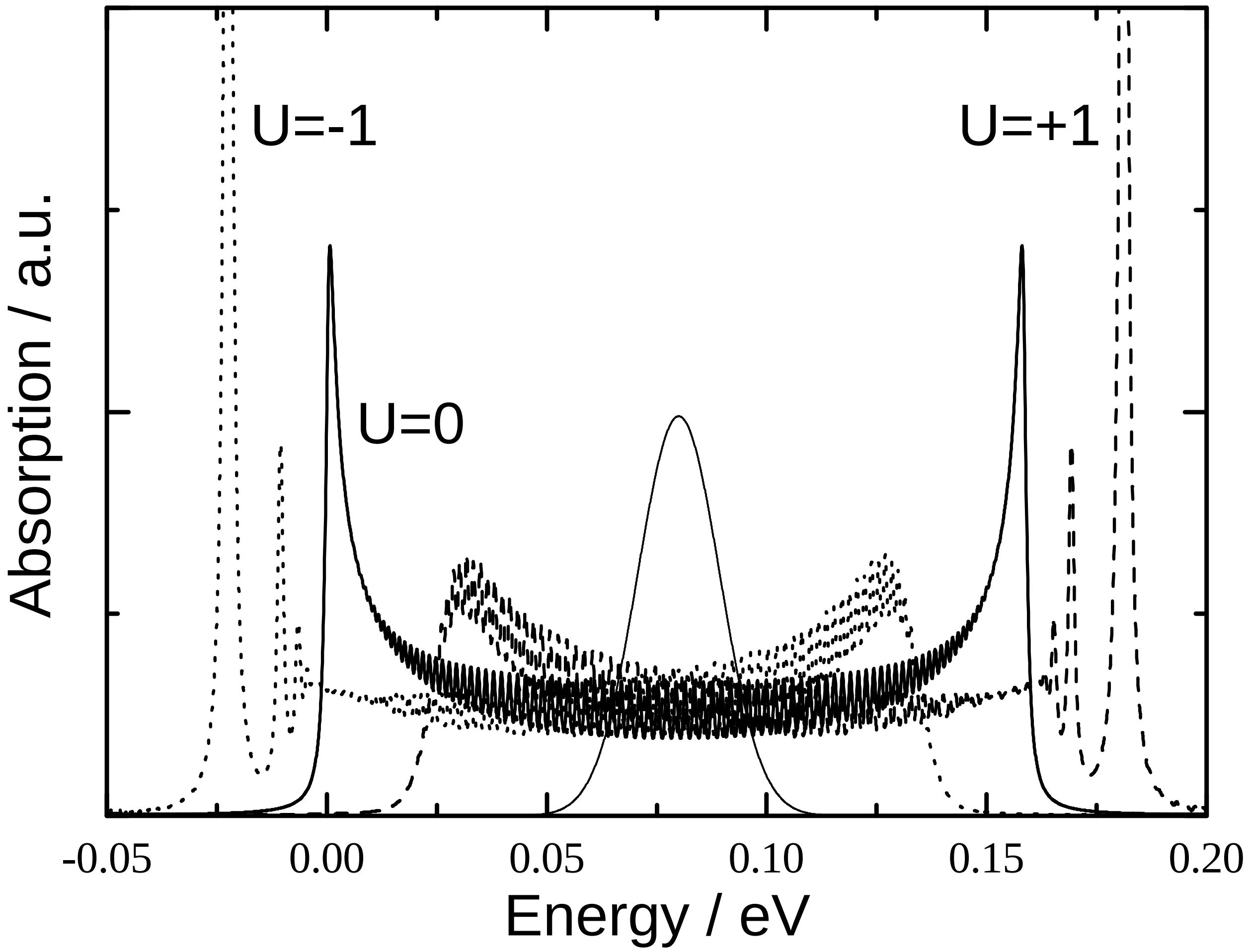

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Figure 1

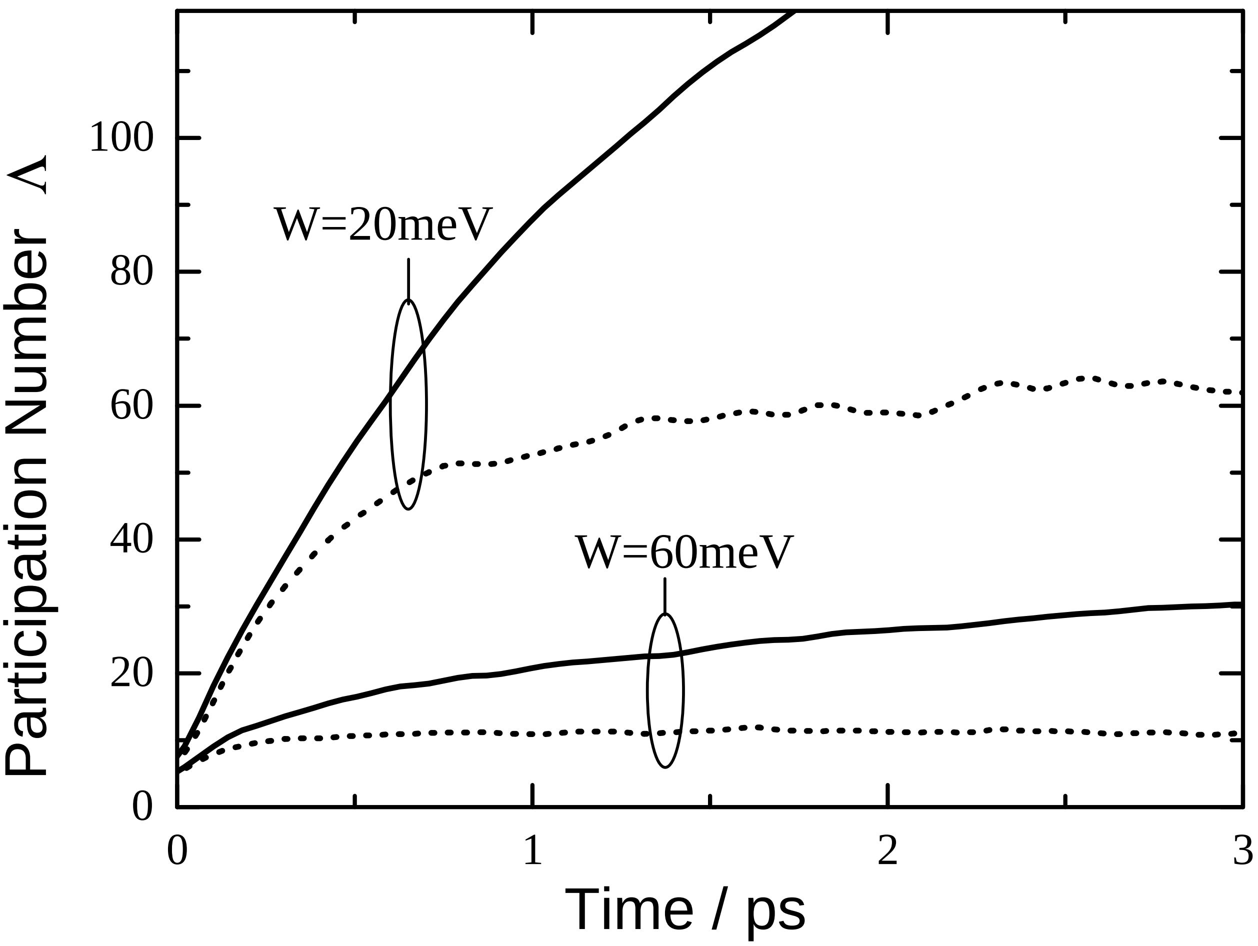

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Figure 2

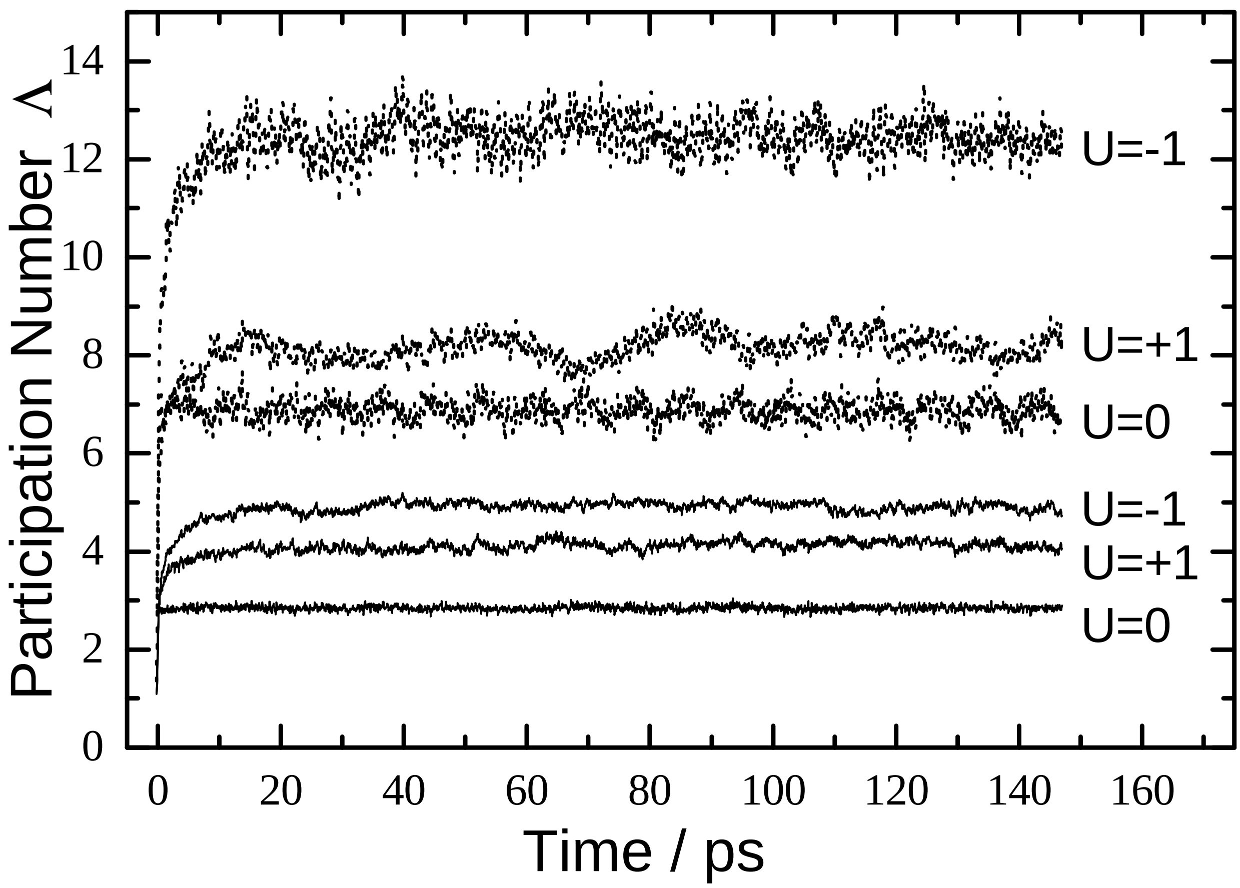

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Figure 3